

Interactive comment on “Lake spray aerosol generation: A method for producing representative particles from freshwater wave breaking” by Nathaniel W. May et al.

Anonymous Referee #2

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* General comments

This paper investigates Lake Spray Aerosol (LSA) through measurements of the size distributions and concentrations of bubbles and aerosol particles produced in an laboratory bubble generator. The measurements are interesting, comprehensive and generally well presented. The manuscript is nicely structured and written. The design of the LSA generator is nothing new. It is a standard plunging jet type bubble chamber. Instead the novelty of the work lies in the combined investigation of bubbles and aerosols generated in fresh- and salt-water solutions. I support publication in ACP after the following comments have been addressed.

The differences in the concentrations of aerosols produced from fresh and saltwater

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seems to be mostly explained by the corresponding differences in bubble concentrations. This is an interesting point, but as the authors have summarised nicely, the increase in the concentration of smaller bubbles in concentrated saltwater solutions due to the inhibition of bubble coalescence was first identified decades ago, and has since been observed in many different studies. I think there is an opportunity to use the combined dataset of aerosol and bubble size distributions to move beyond this point. For example, I suggest adding a plot (or plots) of aerosol mass concentration normalized by bubble concentration (or total bubble surface area or volume) against solution concentration. Fig. 1 indicates that the mass concentration of inorganic ions is $\sim 1000\times$ less in fresh water than salt water. So it is surprising to observe that the total amount of aerosol produced from fresh water in the LSA generator is roughly comparable to the amount of aerosol produced from seawater (as far as I can tell from Fig. 4 taking the different bubble concentrations into account). How is it possible to account for this enormous discrepancy? The authors provide some possible answers in Section 3.2.3 in their discussion of droplet size distributions. Perhaps investigation of normalized aerosol mass concentrations will allow such discussion to be even more quantitative (i.e. the freshwater droplets would need to have $\sim 1000\times$ the volume of the saltwater droplets to produce the same amount of aerosol mass).

The citation and discussion of previous relevant studies is well done. I suggest to also add discussion of the early work of Monahan on freshwater whitecaps (Monahan, 1969; Monahan, 1971). Monahan observed that the wind speed threshold for freshwater whitecap formation is greater than the corresponding threshold for saltwater whitecap formation. He also observed that freshwater whitecaps decay faster than saltwater whitecaps. All things equal, these effects will result in less aerosol production from freshwater than saltwater. The broader point here, which is not made clear in the current introduction, is that the wind speed-whitecap relationship will likely be different over bodies of freshwater than seawater.

****** Specific comments

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P2, L7: 'Larger bubble size distribution' is ambiguous (large sizes or concentrations?).

P3, L12: I would like to see some actual numbers from the Chung et al., (2011) study quoted here to indicate how much LSA might contribute to CCN concentrations. I think its important to state that even using an SSA source function, which should predict greater production of aerosol than a corresponding LSA function according to the results presented here, Chung et al., calculated that LSA only made minor contributions to total particle number concentrations. I think the manuscript overstates the climatic importance of LSA.

P3, L23: The wind speed-whitecap (or -aerosol production) relationship will likely be different for bodies of freshwater than over the oceans. The fetch over lakes is smaller than over the oceans. The threshold wind speed for whitecap formation is greater over fresh water than salt water (Monahan, 1969).

P5, L14: The generator is fine for the purposes of this study, but I think its too much to claim that the generator has been optimized for freshwater experiments. The only feature cited to support this claim is the small volume of the chamber, which is neither novel nor a unique aspect of freshwater experiments.

P7, L18: Can the authors cite any values for typical depth and size of breaking wave bubble plumes? It is important that the depth of the tank doesn't affect bubble plume depth and lifetime, but it is also important to know how the size of the plume in the LSA generator compares to real breaking wave bubble plumes.

P9, L15: Please provide a reference for these known quantification issues.

P9, L18: What value(s) of effective density was used for this conversion?

P11, L6: The concentration of larger bubbles is an order of magnitude less than the Prather et al., (2013) measurements shown in Fig. 3, which are comparable to ocean wave measurements. Even if the decrease in the concentration of larger bubbles follows a similar power law as discussed on P12, L11, the absolute concentrations are

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still very small. This is a limitation of the LSA generator given the importance of larger bubbles as discussed in the next paragraph beginning on L8. This doesn't change the paper's conclusions, the measurements are all internally consistent, but I think the fact should be pointed out.

P16, L12: Is it possible smaller bubbles were present even if they weren't measured?

P22, L9: Another interesting reference to add here is Woodcock's observations from 1948 (Woodcock, 1948).

** References

Monahan, E. C.: Fresh Water Whitecaps, *Journal of the Atmospheric Sciences*, 26(5), 1026–1029, 1969.

Monahan, E. C.: Oceanic Whitecaps, *J. Phys. Oceanogr.*, 1(2), 139–144, 1971.

Woodcock, A. H.: Note concerning human respiratory irritation associated with high concentrations of plankton and mass mortality of marine organisms, *Journal of Marine Research*, 7, 56–62, 1948.

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